

Acid-Dissolution Front-End Process for Mo-99 Recovery at Ambient Pressure: Final Design and Results from Full-Scale Tests

Chemical Sciences and Engineering Division

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Acid-Dissolution Front-End Process for Mo-99 Recovery at Ambient Pressure: Final Design and Results from Full-Scale Tests

by

James Jerden, James Bailey, and George F. Vandegrift
Chemical Sciences and Engineering Division, Argonne National Laboratory

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ACID-DISSOLUTION FRONT-END PROCESS FOR MO-99 RECOVERY AT AMBIENT PRESSURE: FINAL DESIGN AND RESULTS FROM FULL-SCALE TESTS

1 INTRODUCTION

The Global Threat Reduction Initiative (GTRI)-Conversion Program is developing technologies for the conversion of civilian facilities using high enriched uranium (HEU) to low enriched uranium (LEU) fuels and targets. The conversion of conventional HEU targets to LEU for ^{99}Mo production requires approximately five times the uranium in a target to maintain the ^{99}Mo yield on a per target basis. As part of the GTRI-Conversion Program, Argonne has developed two front-end options that allow the use of LEU-foil targets in current schemes for ^{99}Mo production.

The two front-end processes developed as part of this project are (1) the dissolution of irradiated LEU foil (up to 250 g in a single batch) and nickel fission recoil barrier in nitric acid at ambient pressure followed by separation of Mo on a titania column and (2) the electrochemical dissolution of LEU foil. Both these processes produce an alkaline (basic) solution feed for ^{99}Mo purification. This report describes results from full-scale performance tests of a nitric-acid-dissolver system that operates at ambient pressure. The system is designed for dissolution of up to 250 g of irradiated LEU foil and associated fission recoil barrier metal (e.g., Ni). The design, fabrication, and previous performance tests for this system are described in more detail in earlier reports [Jerden et al., 2011, 2013]. A separate report discusses the chromatographic separation of Mo from the acid solution and its recovery in alkaline solution [Stepinski et al., 2014, in preparation].

This report gives an overview of the final dissolver system design and results from three new performance tests, including a full-scale experiment using slightly irradiated LEU foil.

2 NITRIC-ACID-DISSOLVER SYSTEM FOR LEU FOILS

Using irradiated and non-irradiated foils, we tested the nitric-acid-dissolver system at full scale. The key design criteria of the acid dissolver system are summarized below:

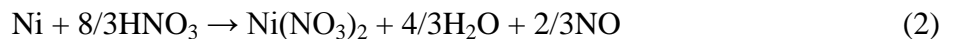
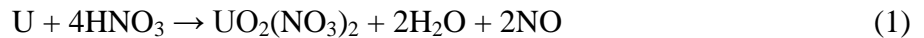
- All water vapor, reaction products, and fission gases are contained within the dissolver system at a maximum temperature of 125°C and 2 atm of pressure (absolute) under both normal and off-normal (loss of cooling during reaction) conditions.
- The acid-feed system is designed so that the thermally hot LEU foil (hot from decay heat) can be immersed in nitric acid without losing solution due to instantaneous boiling.
- Gas-trap components are designed to trap/neutralize all nitrogen oxide and acid gases (NO, NO₂, HNO₂, and HNO₃) as well as trap iodine gas.
- Noble fission gases are passively contained within the system.
- All dissolver system components are designed for remote operation in a hot cell facility.

This last design feature involved direct, hands-on collaboration with hot-cell operators from the Radiochemical Engineering Development Center (REDC) at Oak Ridge National Laboratory. Figure 1 presents a schematic diagram showing the major components of the LEU-nitric acid front-end process.

Before discussing the full-scale dissolution experiments, we summarize the design basis requirements for the dissolver system.

2.1 DISSOLUTION REACTION: REACTION HEAT, DECAY HEAT, AND OFF-GAS VOLUME

The volume and concentration of nitric acid used for a given experiment depend on the mass of the metal being dissolved as well as the desired final acid concentration of the “product” solution (i.e., the solution produced by the dissolution experiment). Controlling the final acid concentration is important for optimizing the Mo-99 extraction step, which follows the dissolution step. The volumes and concentrations of acid as well as the amount of nitrogen oxide gas (NO_x: NO, NO₂, and N₂O₄) that will be produced are determined by the following general reactions:



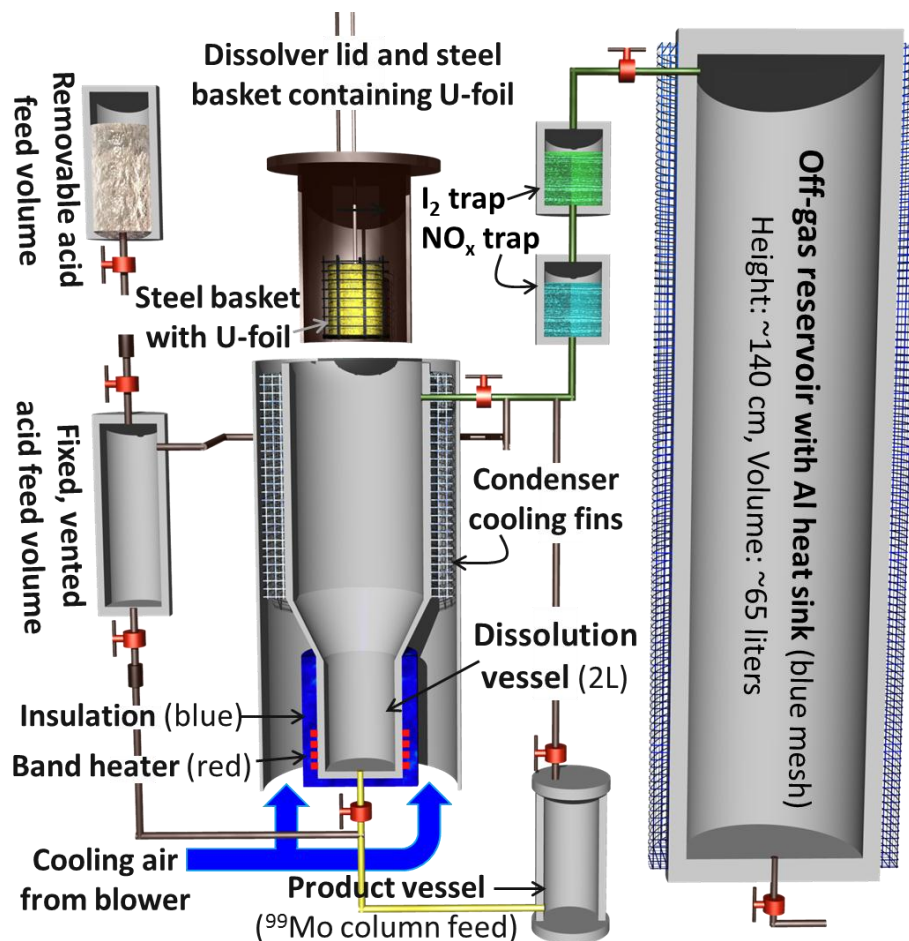


FIGURE 1 Schematic Cross Section Showing Components of the LEU Nitric-Acid-Dissolver System Designed and Tested at Argonne National Laboratory

In the presence of oxygen, the $\text{NO}(\text{g})$ produced in these dissolution reactions is rapidly converted to $\text{NO}_2(\text{g})$:



When water vapor and oxygen are present, NO_2 is readily converted to both nitrous and nitric acid vapors [$\text{HNO}_2(\text{g})$ and $\text{HNO}_3(\text{g})$], which will dissolve in condensed water and flow back down into the dissolver.

One of the main purposes of the experiments discussed in this report is to determine how much acidity is lost from the dissolver solution as a result of the escape of NO_x and acid gases from the condenser section of the dissolver.

Most of the off-gas from the dissolver will consist of NO_x [Jerden et al., 2011]; however, ORIGIN calculations show that iodine, xenon, and krypton will also be present in the off-gas.

The ORIGIN calculations (performed by Charlie Allen, University of Missouri, using ORIGIN2, Version 2.2) assume the following:

- Irradiation of 1 g of uranium foil enriched to 19.75% ^{235}U ,
- Power = 1.9×10^{-3} MW,
- Burnup = 1.59×10^{-2} MW-days,
- Flux = 2.1×10^{14} N/cm²-sec, and
- Burnup time = 200 hr.

The foil composition is given for cooling times of 12, 24, 36, and 48 hours. The most abundant off-gas species will be iodine (4.2×10^4 Ci per 250 g LEU after 12-hr cooling), xenon (2.6×10^4 Ci per 250 g LEU after 12-hr cooling), and krypton (1.7×10^3 Ci per 250 g LEU after 12-hr cooling).

Thermodynamic calculations show that for the dissolution of 250 g uranium metal (~1.05 moles), the total energy released may be up to 1600 kJ but will probably be closer to 1000 kJ. If we assume adiabatic conditions and a 30-min reaction time, this energy would correspond to a maximum thermal power output of approximately 890 W [Jerden et al., 2011]. The ORIGIN calculations show that a total thermal output for 250 g of irradiated LEU is around 1000 W for a 12-hour cooling (Figure 2).

Based on the heat of dissolution and decay heat calculations, the cooling system for the LEU nitric-acid-dissolver system must be able to sink out a maximum of 2000 W (thermal). Therefore, if we assume that if the dissolution of 250 g of irradiated LEU foil takes 30 min, 2000 W of thermal power will be generated and will need to be removed from the system to ensure that water vapor (and acid) is not lost during the dissolution. To test the performance of the dissolver cooling system, we conducted experiments in which both the reaction and decay heats were simulated by using heating coils wrapped around the dissolver vessel.

2.2 DESIGN CRITERIA FOR DISSOLVER CONDENSER SECTION AND OFF-GAS RESERVOIR

A mathematical model was implemented in Mathcad for the design and design optimization of the nitric-acid-dissolver system with LEU foil. This Mathcad model is supported by heat flow calculations performed with the ANSYS CFX code.

The current dissolver design was based on the Mathcad/ANSYS CFX model. An example of model results for the off-gas reservoir is shown in Figure 3. By comparing the model calculations with test results (discussed below), we have optimized the system design: specifically, the size of the cooling system fan and design of the off-gas reservoir. The Mathcad model was used for the following purposes:

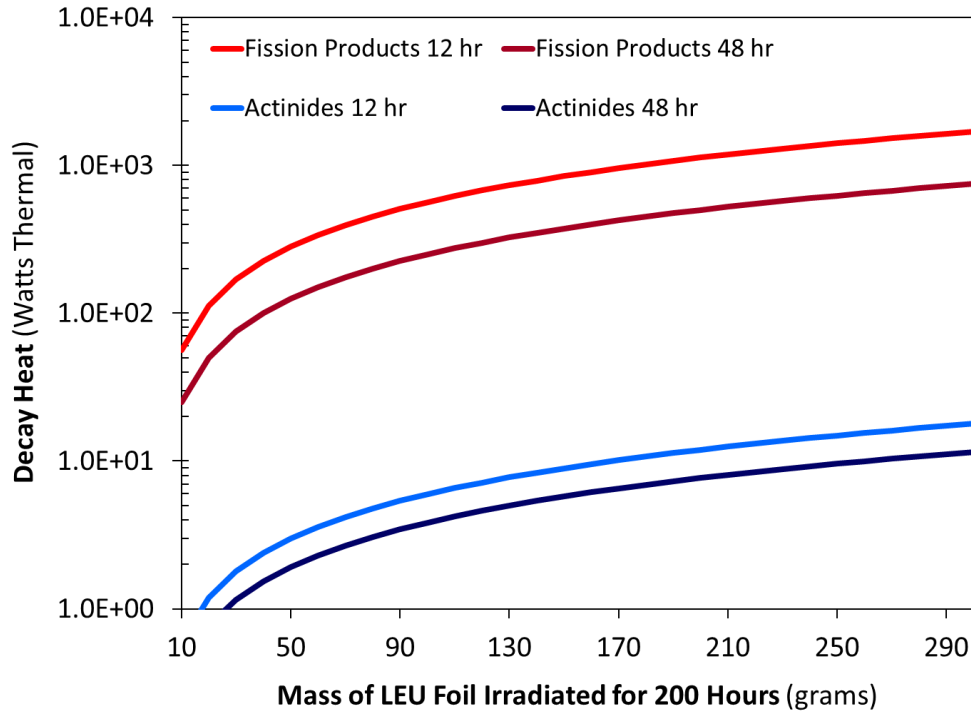


FIGURE 2 Cumulative Decay Heat for Fission Products and Actinides for a Range of LEU Foil Masses after 200-Hour Irradiation. Curves are for 12 and 48 hours of cooling (time out of reactor). Data calculated by using ORIGEN2.

- Sizing of the cooling fins and air flow requirements from the cooling fan to remove both the reaction heat and decay heat from the dissolver. With a reaction heat of approximately 1600 kJ over 30 min and a constant decay heat of approximately 1500 W, the average required heat removal rate is 10181 kJ/hr. This value determined the original cooling system design.
- Calculation of the pressure drop across the cooling fin section based on the required flow velocity as determined by the convective heat removal from the fins. Also calculated was the total air volume flow rate and temperature rise of the air across the fins. These results were used to determine the cooling fan size from fan performance curves.
- Calculation of the required gas volume and heat sink capacity of the off-gas reservoir. We assumed that all the reaction heat and off-gas are transferred to the reservoir (loss of cooling scenario). Also, the steady-state requirement for the decay heat removal was assumed to be dissipated by the reservoir. A thermal analysis was performed to determine an approximate value for the heat loss from the off-gas reservoir. The reservoir tank was assumed to be maintained at a uniform temperature of 100°C with an ambient air temperature of 32°C. Heat losses due to radiation and natural convection are calculated

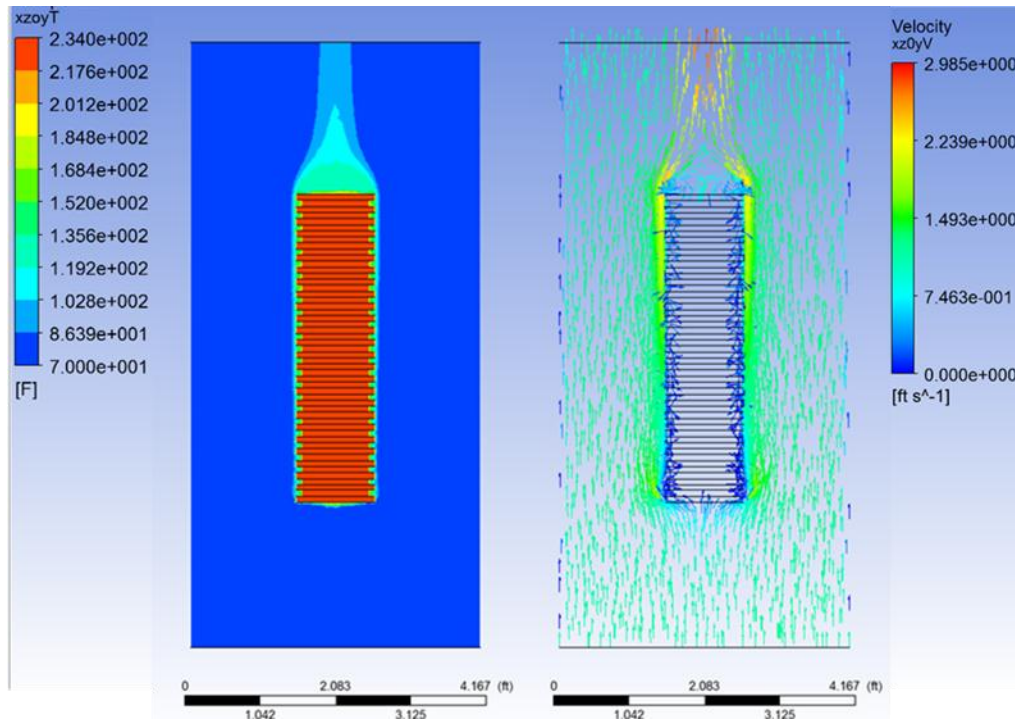


FIGURE 3 Example of Results from ANSYS CFX Model for the Off-Gas Reservoir for the Dissolver System. Left: Surface temperature (red) of dissolver reservoir with 1,500 W internal heat generation. Right: Natural convection velocity around the exterior of dissolver reservoir. Calculations show that the heat sink (cooling fins) attached to the reservoir is sufficient to keep the system from becoming pressurized in the event of a complete loss of active cooling.

separately and then added together to determine the total heat flow from the outside of the reservoir to the air in the hot cell. Results from this calculation were used to size and design the aluminum heat sink rings that encase the off-gas reservoir tank.

- Calculation of the heat-removal capacity due to condensing of the process vapor on the inside surface of the dissolver condenser section. These results were accounted for in the cooling system design. Boiling of the dissolver solution occurs in the vessel at the bottom of the dissolver, and the vapor moves upward to the condenser section, where it condenses on the cooler walls that have been cooled by the air flow over the fins on the outer surface. The wall temperature is assumed to be below 100°C.

2.3 DISSOLVER SPECIFICATIONS

The dissolver system design has been updated a number of times in collaboration with hot-cell operators at Argonne and Oak Ridge National Laboratory to ensure that the final design

is easily operated using typical manipulators. For design details, see previous reports [Jerden et al., 2011, 2013]. The final design, footprint, and a photograph of the complete dissolver system are shown in Figure 4. Details of the final design and a photograph of the NO_x and iodine gas traps are shown in Figure 5. Figure 6 shows photographs of the dissolver vessel and condenser section in various stages of preparation for the dissolution experiments, including the locations of thermocouples for monitoring thermal gradients during the tests.

The dissolver vessel is open to the off-gas reservoir during the dissolution process. The volume of the reservoir and the sizing of the aluminum heat sink were chosen to provide passive containment of all water vapor and reaction products at a pressure less than 2 atm (absolute) during both normal and off-normal (loss of cooling during reaction) conditions.

A step-by-step summary of the operation of the dissolver system is shown in Figure 7. The dissolution process is started by first lowering the uranium foil (contained within a steel mesh basket) into the dissolver vessel and then sealing the vessel with a metal cap. Nitric acid is then added to the vessel using a two-chamber acid feed system that is designed to avoid pressurization of the acid bottle in the event that the dissolution reaction begins instantaneously when the acid is added. The dissolver vessel is cooled by forced air blown from the base of the unit. The temperature of the dissolver solution is monitored by a thermocouple. The dissolver vessel is insulated so that the top of the vessel is cooled continuously during the reaction. Heat loss from the top of the vessel is optimized by the presence of steel cooling fins attached to the condenser part of the dissolver system. This design causes the water and acid vapors to condense along the walls at the top of the vessel during the dissolution reaction (as acid is boiling). The role that the condenser section of the dissolver plays during a typical dissolution run is shown schematically in Figure 8.

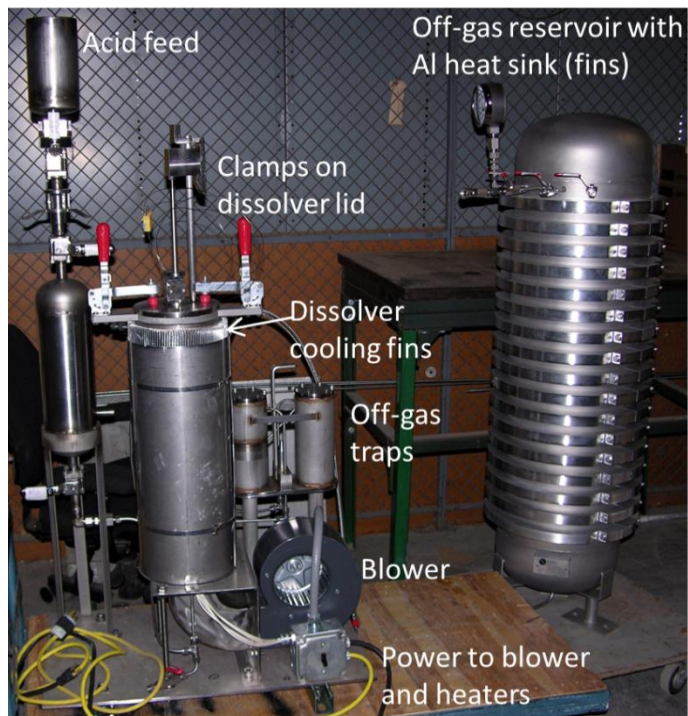
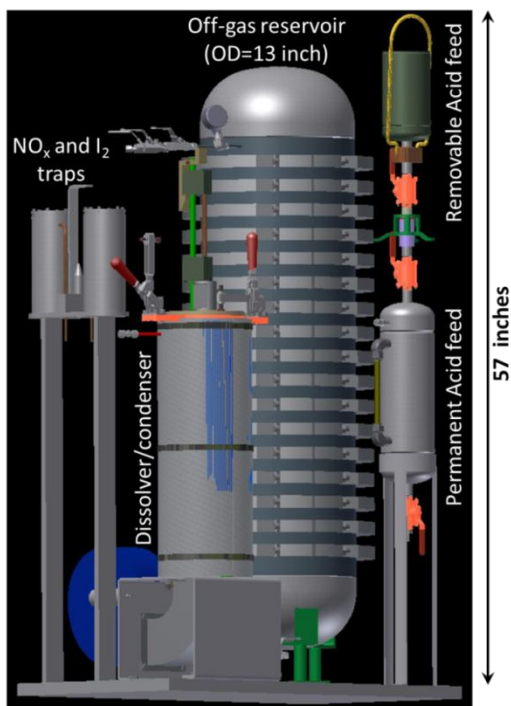
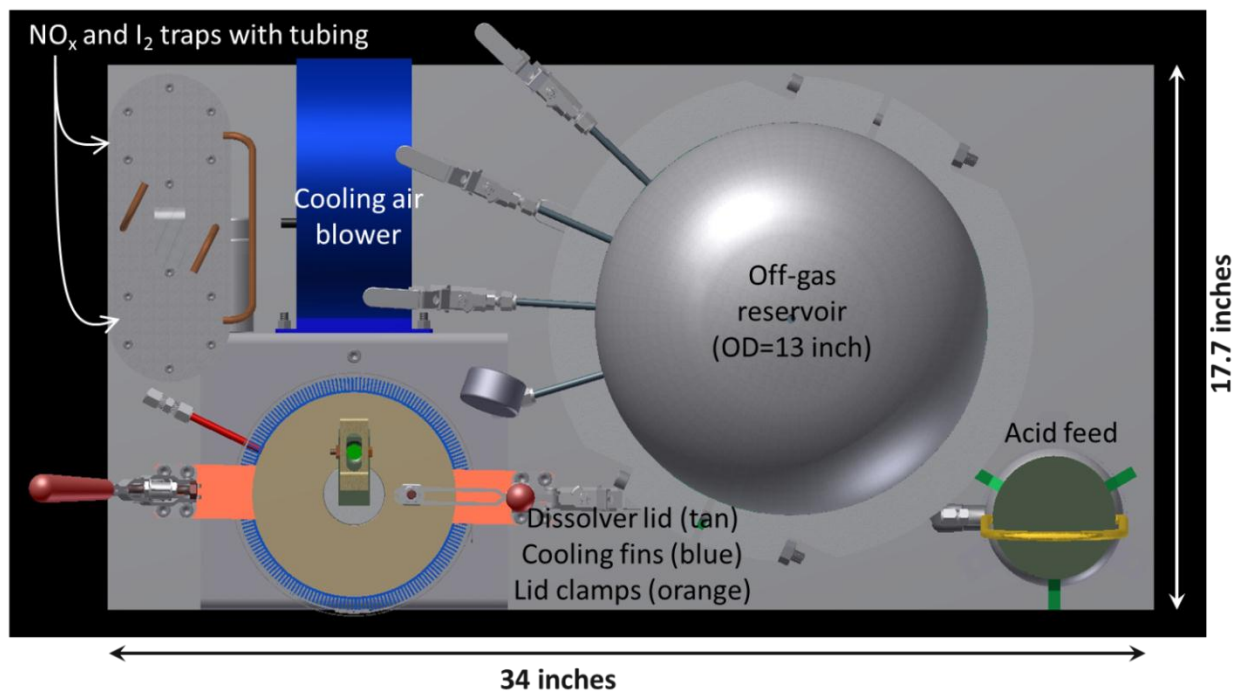


FIGURE 4 Drawings and a Photograph of the Dissolver System for the Completed Final Layout Showing the Key Dimensions for Setting Up the System in a Hot Cell

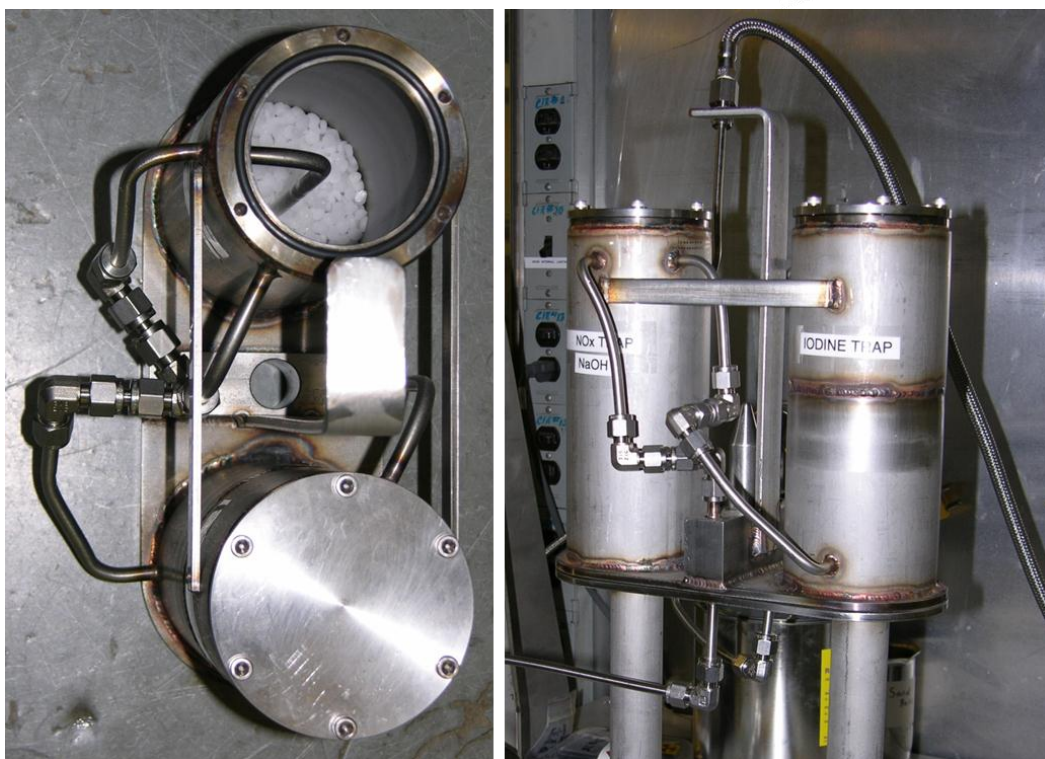
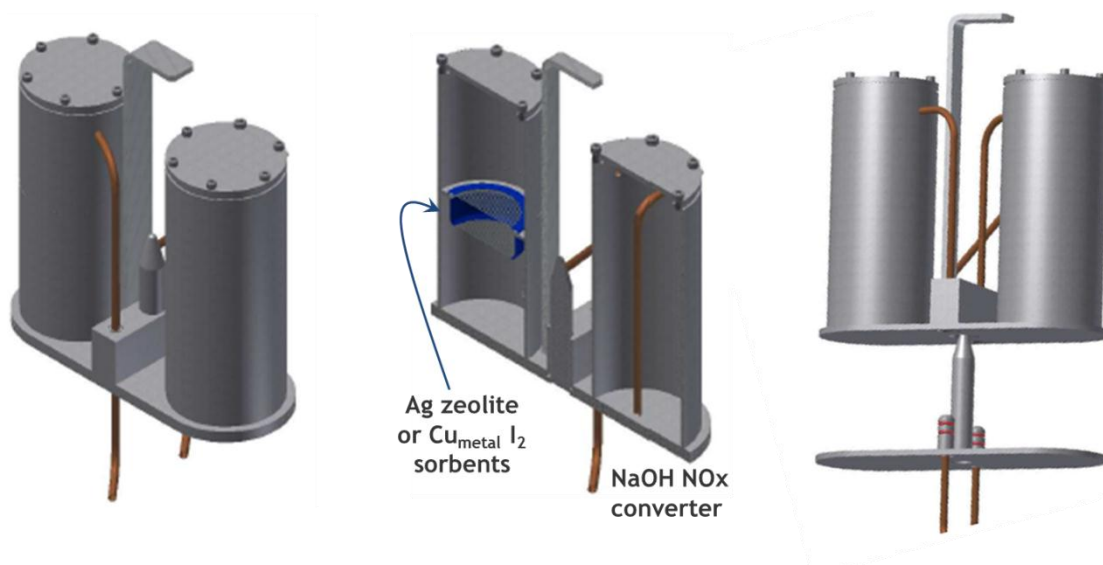


FIGURE 5 Drawings and Photographs of the Off-Gas Traps Used for Performance Tests

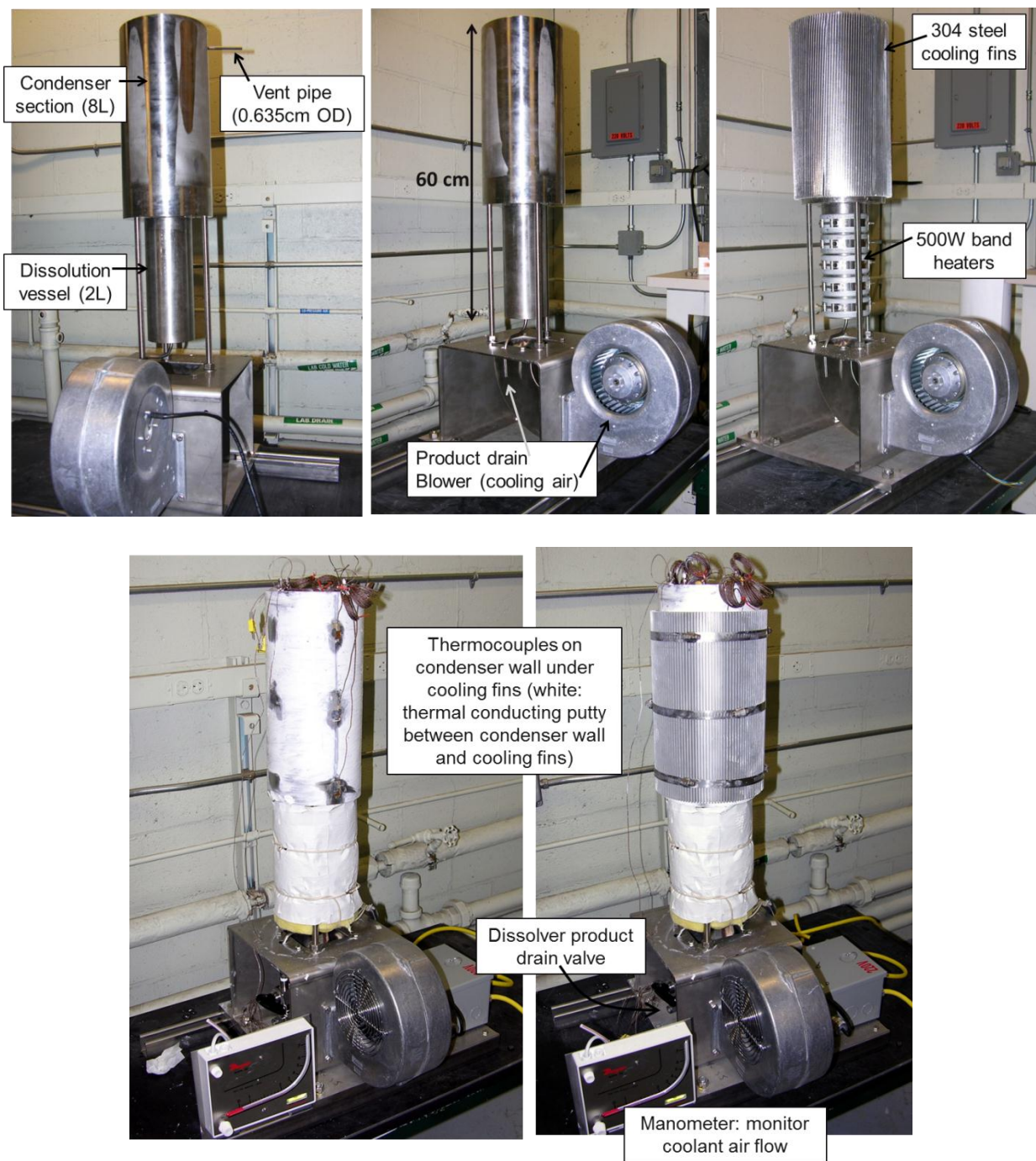


FIGURE 6 Dissolver/Condenser and Cooling Fan Portion of Full-Scale Dissolver Prototype Used in Performance Tests

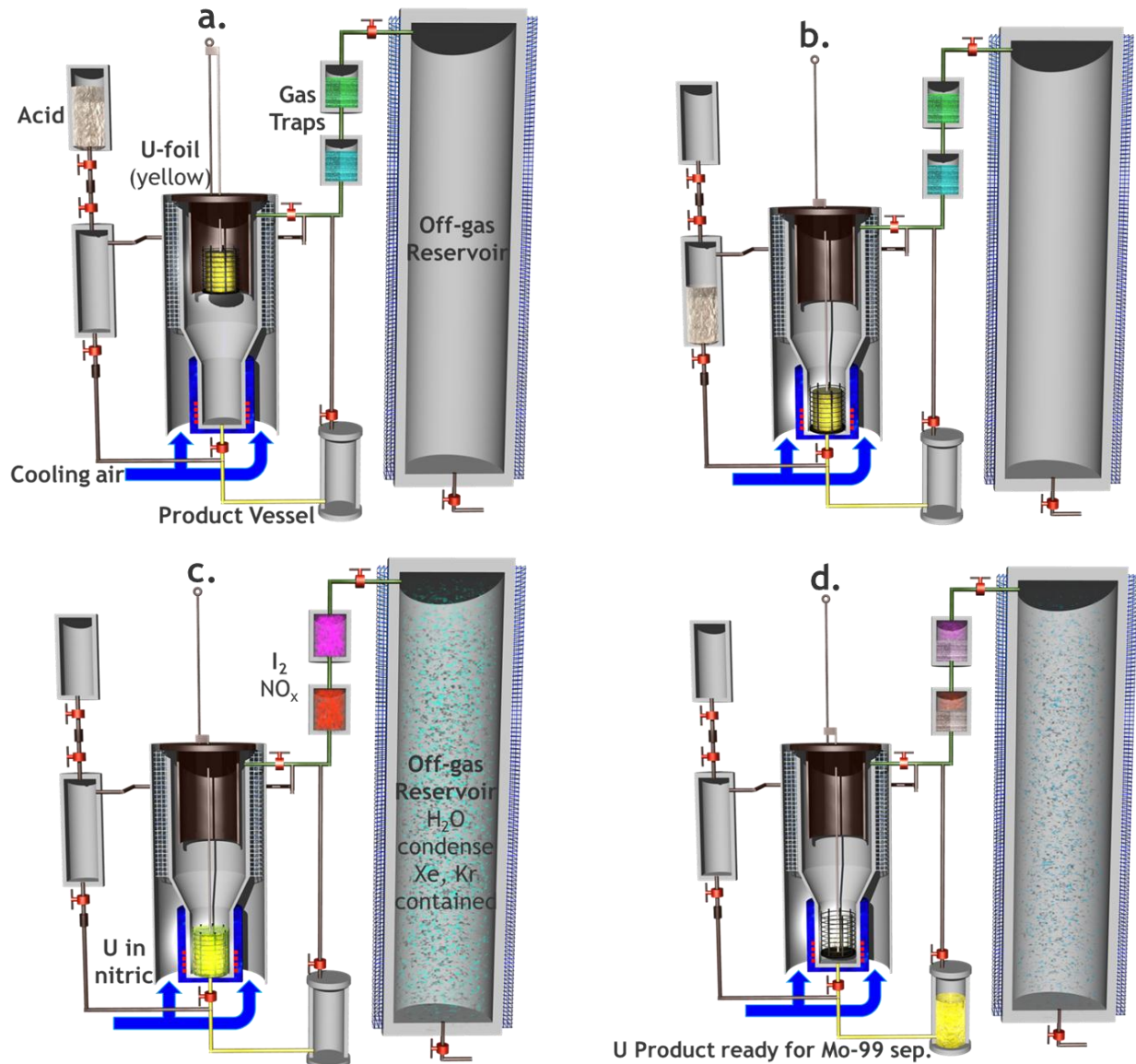


FIGURE 7 Conceptual Diagrams Showing the Four Main Operation Steps with the Nitric Acid Dissolver System Optimized for the Hot Cell Space. (a) Fill removable acid feed vessel and load irradiated foil into steel basket, (b) drain acid into permanent, vented acid feed volume (close valves) and lower irradiated foil basket into dissolution vessel, (c) drain acid into dissolution vessel to start foil digestion, where off-gas reacts with gas sorbents (NO_x shown in orange; I₂ as magenta; Xe, Kr as blue), and (d) drain product of foil digestion into vented vessel, which will serve as the feed for the Mo-99 extraction columns.

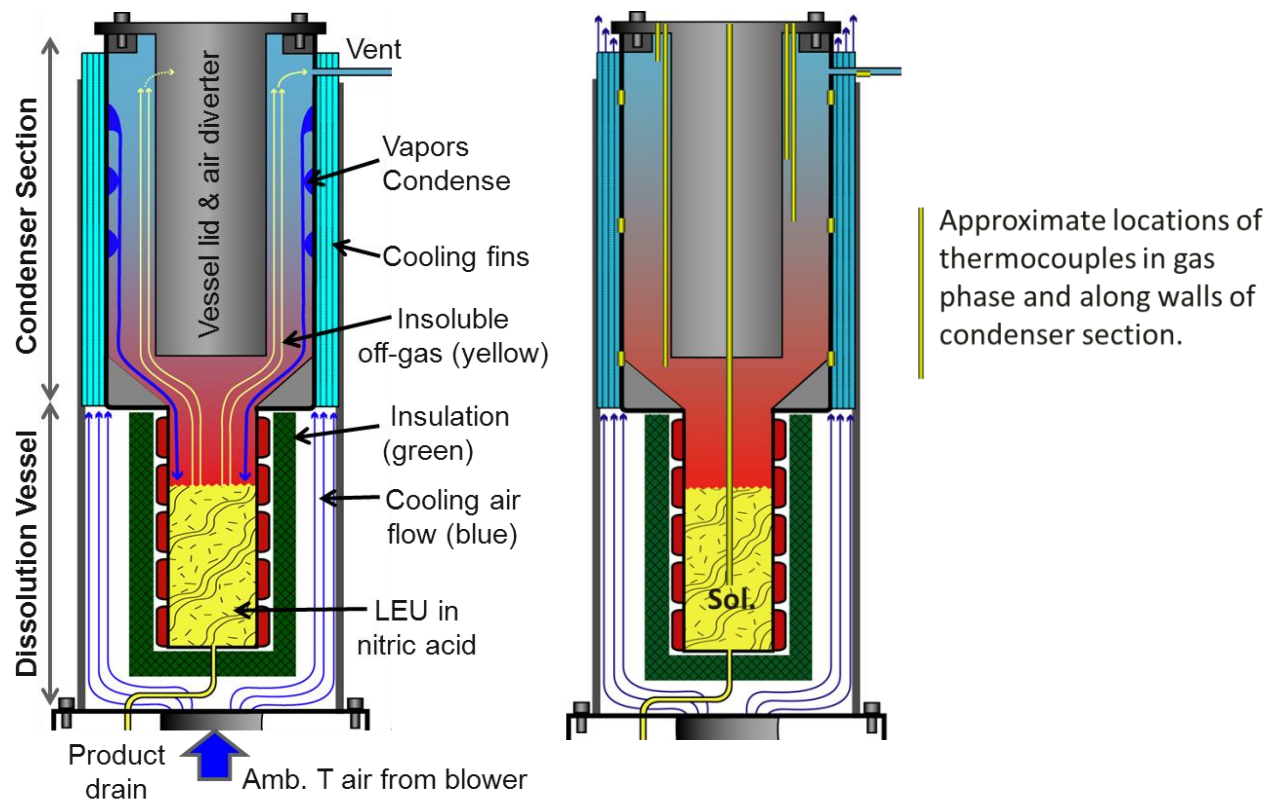


FIGURE 8 Schematic Cross Sections through Dissolver Vessel and Condenser Section Summarizing Processes during the Foil Dissolution Process

3 DISSOLVER SYSTEM PERFORMANCE TESTS

3.1 METHODS

Three experiments were performed. First, 197.26 g of depleted uranium was dissolved discontinuously, that is, the test was interrupted to determine the extent of dissolution after 20, 40, 80, and 90 min. The acid dissolver solution was not changed out during the interruptions of this test. Second, 202.02 g of depleted uranium was dissolved with minimal heat input to measure the temperature profile produced by the heat of dissolution of the foil. Third, 242.4 g of depleted uranium foil was combined with 6.84 g of irradiated LEU foil and dissolved with the full heat load.

Nickel metal was added to each test in an amount consistent with its presence as a 7–10 μm - thick fission recoil barrier in the actual target. For the dissolution experiments discussed here we assumed that 4% of the mass of a given uranium foil target is nickel (based on the relative thicknesses of the metals). Therefore, we added 10 g of nickel foil to the 197 and 202 g U tests and 13 g of nickel to the 242 g U test.

Based on the stoichiometry in reactions (1) and (2) and an acid volume of 500 mL, the starting acid concentrations for the three tests would be around 8.7 M nitric for the 197 g and 202 g U tests and 10.5 M nitric for the 242 g U test to achieve a final acid concentration of 1 M (optimal for Mo-99 extraction on titania column). However, it is assumed that the dissolution will not follow reactions (1) and (2) due to the loss of NO_x gas through the gas traps. Therefore, excess initial acid was used. For the 197 g and 202 g U tests, the initial acid concentration was 9.7 M, and for the 242 g U test, it was 11.5 M. The experimental setup for these test is shown in Figure 9, and example photographs of the dissolver basket and the foils used for Test 3 are shown in Figure 10.

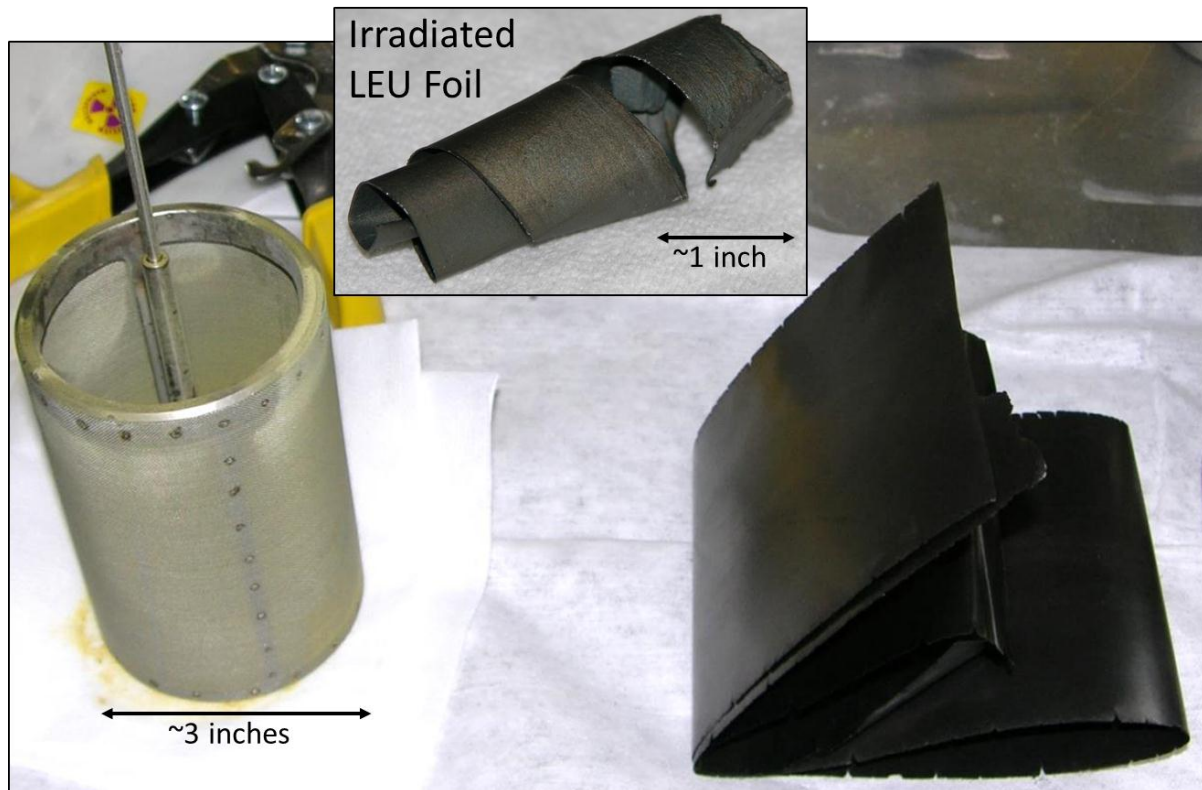
The acidity of the dissolver solution was determined by the potentiometric titration method of Motojima and Izawa [1964]. This method involves mixing an aliquot of the dissolver solution sample (approximately 0.5 mL) with 5 mL of 2 molar ammonium sulfate solution and titrating the mixture to the equivalence point, as identified on the titration curve. The ammonium sulfate complexes uranium to prevent hydrolysis during base titration.

After determination of the acid concentration, the dissolver solution from Test 3 was adjusted to 1 molar HNO_3 and used as the feed for Mo-99 extraction tests employing a titania column (to be discussed in separate report).

The linear flow velocity of the cooling fan was measured throughout each experiment. The cooling air flow was constant at 25.9 m/s where it entered the cooling fin section at the base of the dissolver. The cooling air flow measured at the top of the dissolver, where it exits through the cooling fins, was consistently 20.1 m/s. The loss of flow velocity is due to turbulence as the cooling air travels up through the dissolver duct work and is channeled into the cooling fins and, perhaps, by small leaks in the steel sheath that contains the fins.



FIGURE 9 Full-Scale Dissolver Prototype Used in Uranium Foil Performance Tests



Basket containing DU foils lowered into dissolver vessel

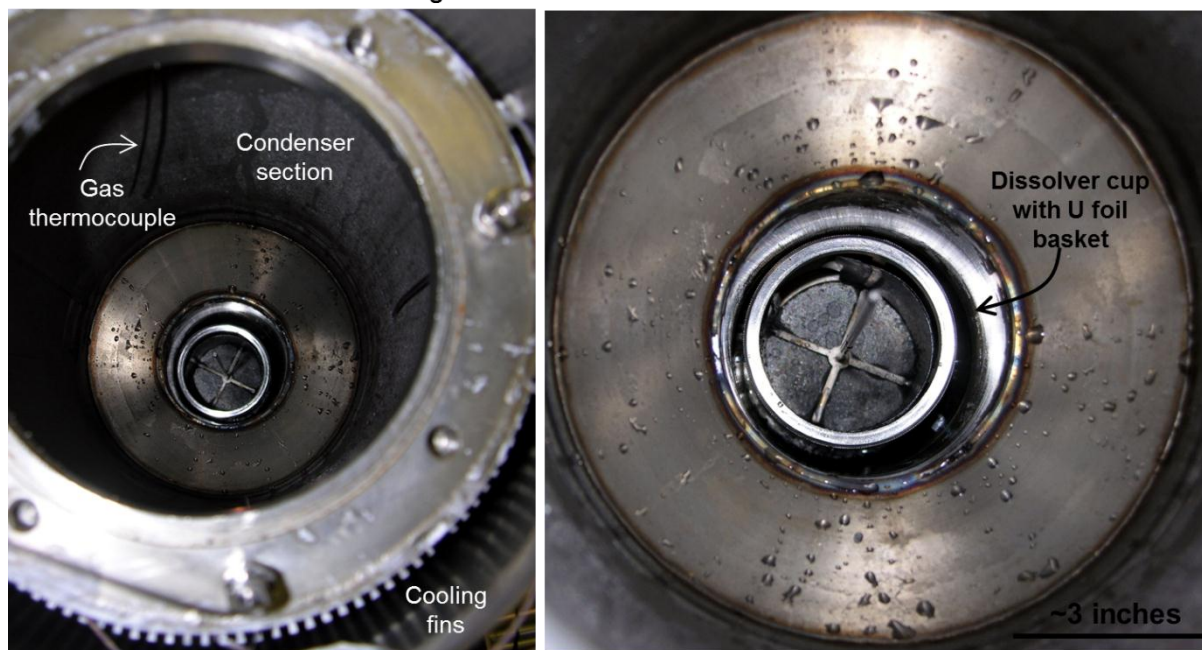


FIGURE 10 Dissolver Basket and the Foils Used for Performance Tests. The top photograph shows the 242.4 g of depleted uranium and the 6.84 g of irradiated LEU foil used in Test 3. The bottom two photographs are from experiment performed last year with 133 g of depleted uranium showing how the foil basket fits in the dissolver cup at the base of the condenser section of the dissolver [Jerden et al., 2013].

3.2 RESULTS AND DISCUSSION

The approximate locations of thermocouples for the dissolver performance tests are shown in Figure 8 (right plot). The temperature data for the minimum heat test (No. 2) and the full-scale test (No. 3) are shown in Figures 11 and 12. There are no temperature data from the discontinuous test (No. 1) due to the interruption of the thermocouple measurements throughout the test. Figures 11 and 12 show the temperature of the dissolver solution, as well as the maximum and minimum temperatures of the condenser section wall and gas phase. Data from the other thermocouples fall between these curves in a predictable manner, that is, the temperatures are higher near the base of the dissolver where the heaters are located.

The heat of dissolution measurement shown in Figure 11 indicates that approximately 225 kJ of heat is liberated from the dissolution reaction in this experiment (assuming a heat capacity of 2.68 J/kg·°C for the acid solution). We cannot compare this number directly to the thermodynamic calculation for the enthalpy of reaction of the fuel dissolution because the dissolver is a poor calorimeter (the gas phase above the solution is being actively cooled).

For the full-scale test (Figure 12) the wattage of the heaters was initially set at 1600 W for the first 50 min and then increased to 2000 W, which simulates the total heat load for a single production-scale run. The thermal data show that the cooling of the condenser section is effective even at the maximum power. The forced air cooling system maintains a thermal gradient of around 30°C between the maximum gas and dissolver wall temperatures for the duration of heating.

Results from the discontinuous dissolution test show that within 20 min of the start time to the test the uranium oxide layer was removed from the foils, as evidenced by a change in color from dark gray to a dark gold (compare Figures 13a and 13b). Also, sharp edges for the foil were dissolved at this stage. Around 40 min after the start of the test, particular sections of the foil were preferentially attacked while other sections hosted uranium solid precipitates, such as uranyl nitrate (top part of Figure 13c). This effect may be due to the poor mixing of the solution in recesses of the foil basket and the establishment of local chemical environments. After 80 min, less than a gram of foil remained at the bottom of the foil basket (Figure 13d). After another 10 min that last bit of foil dissolved. Even after 20 min, there was no sign of the nickel foil that had been added to the basket, indicating relatively rapid dissolution of the nickel foil relative to the uranium.

The starting acid concentrations for these experiments were 9.7 molar for the 197 and 202 g U tests and 11.5 molar for the 242 g U test. These concentrations are in excess of the starting concentrations required to end up with 1.0 molar nitric acid if the dissolution follows the stoichiometry described in reactions (1) and (2). The higher-than-stoichiometric starting acid concentrations were used to counter the loss of acid through the loss of NO_x gas during the tests.

Using the potentiometric titration method of Motojima and Izawa [1964], we found only a minor (if any) decrease in acidity due to the loss of NO_x gas during the dissolution in all three tests. The acidity measurement involved adding a 0.5 mL aliquot to 5 mL of 2 molar ammonium sulfate solution and then titrating with NaOH to an equivalence point. The acidities of the final

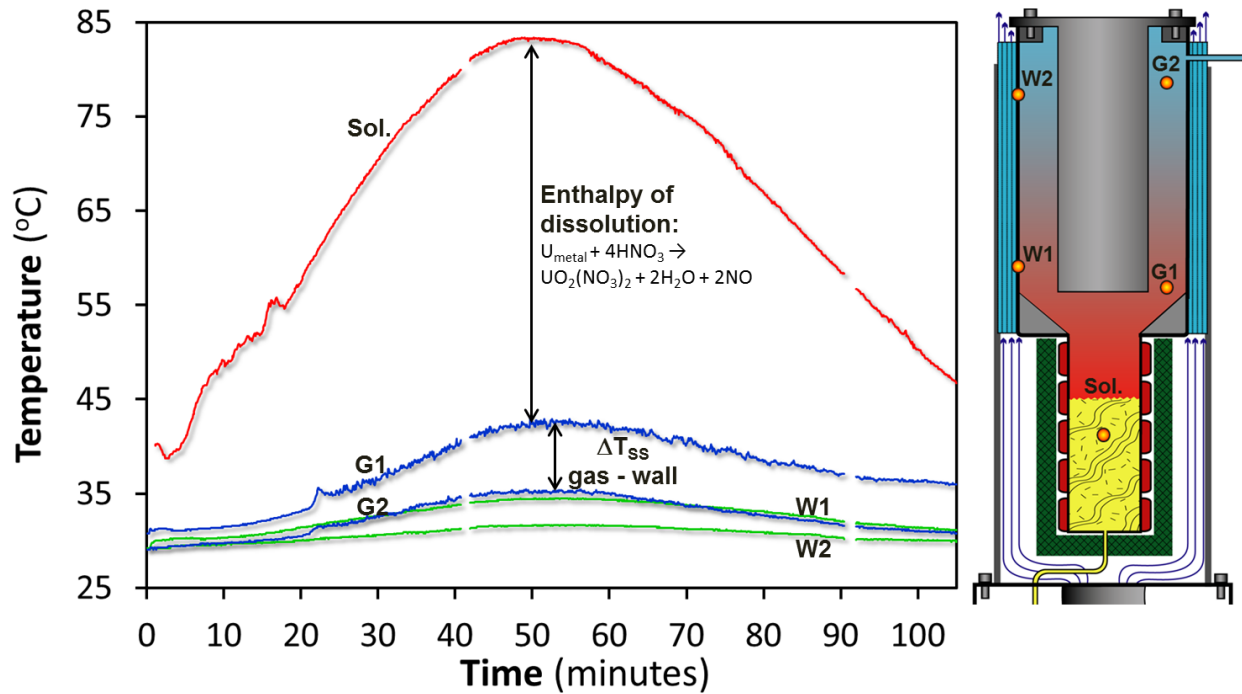


FIGURE 11 Thermal Profiles within the Dissolver and Condenser Sections during Enthalpy of Dissolution Test in which a Small Amount of Heat was Added to Start and End the Dissolution Reaction. The purpose of the test was to determine the heat output from the uranium foil dissolution reaction.

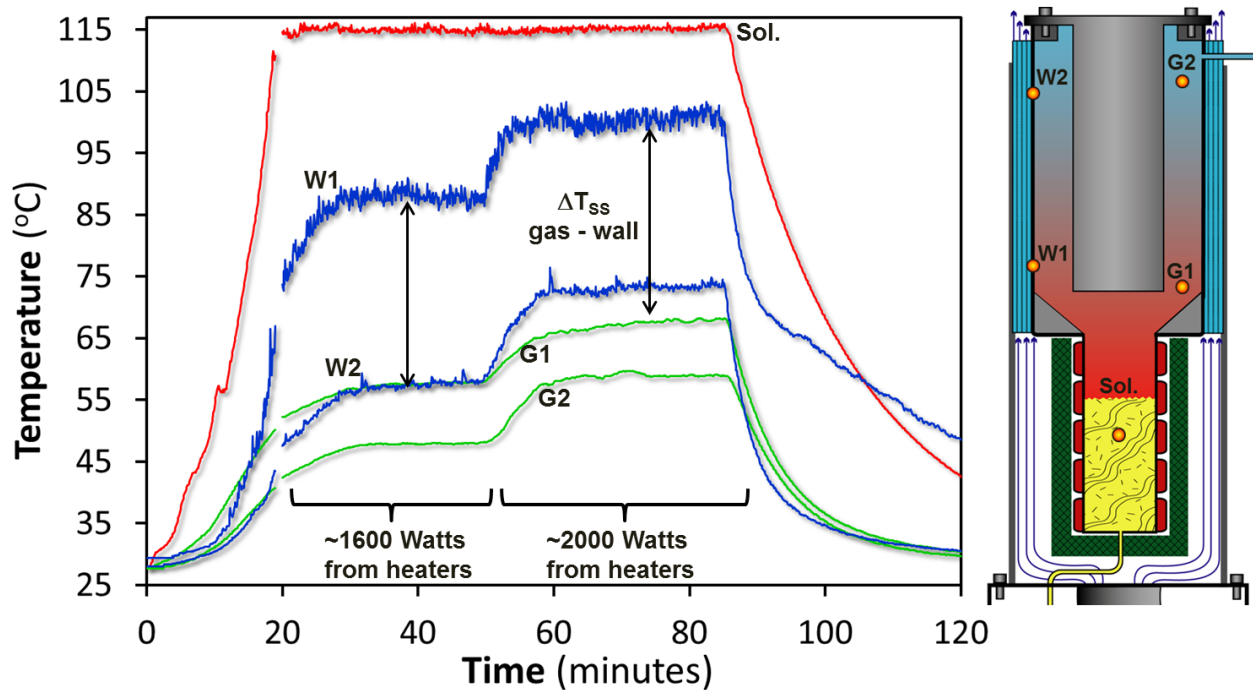


FIGURE 12 Thermal Profiles within the Dissolver and Condenser Sections during Full-Scale Dissolution of 242.4 g of Depleted Uranium and 6.84 g of Irradiated LEU Foil (Test 3)

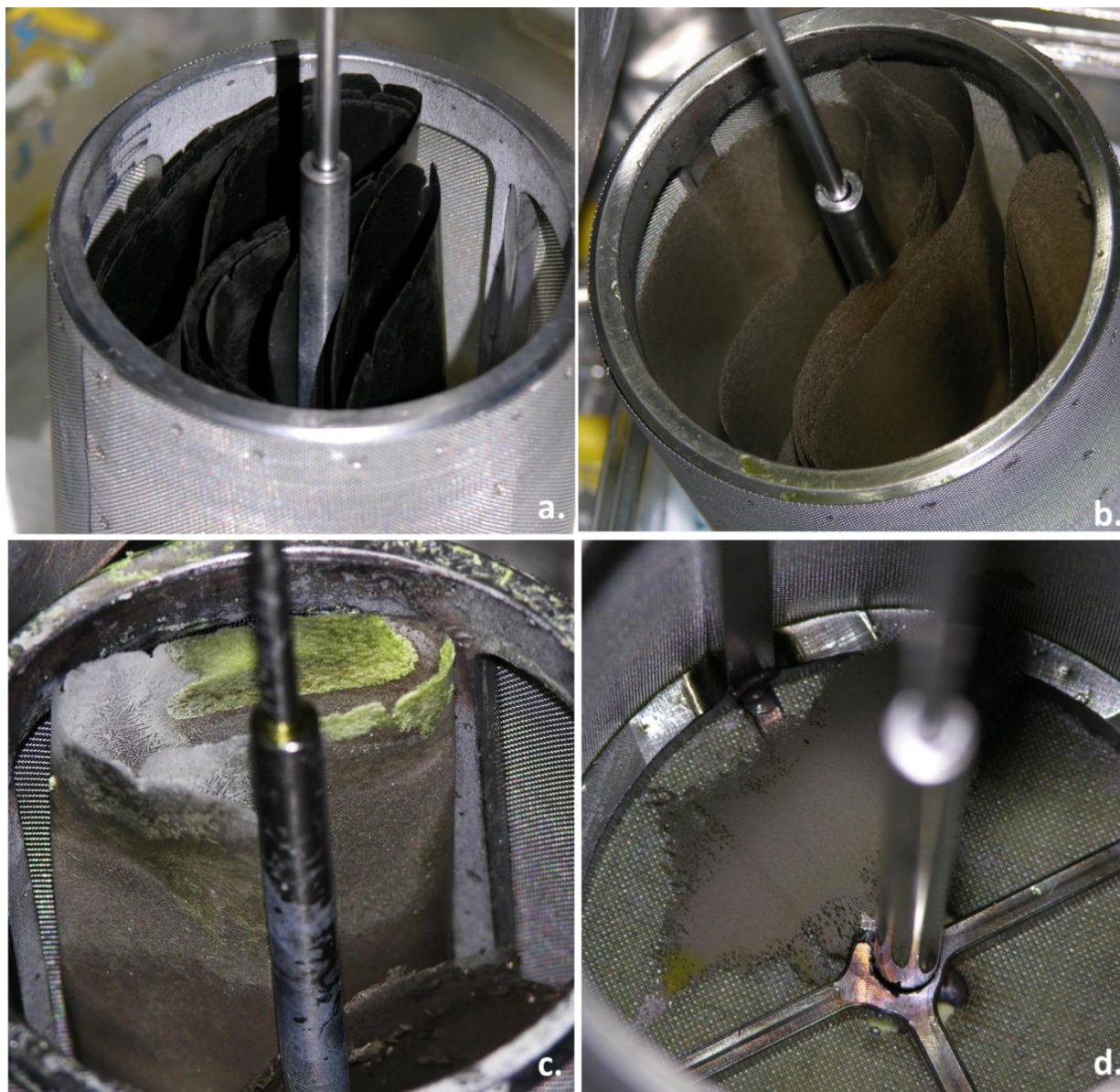


FIGURE 13 Photographs of Depleted Uranium Foil in the Dissolver after (a) 0 min, (b) 20 min, (c) 40 min, and (d) 80 min in Test 1. Note the acid dissolver solution was not changed out during the interruptions of this test.

solutions were as follows: 1.8 M for Test 1, 2.1 M for Test 2, and 2.1 M for Test 3. These results indicate that the dissolution is close to following the stoichiometry shown in reactions (1) and (2). Based on the acidity measurement and the amount of uranium in solution, we adjusted the final solution acidity from Test 3 (250 g U) to 1.0 M by adding NaOH prior to transferring the resulting solution to the Mo-99 extraction step.

We also observed a significant amount of NO_x gas being released while the final solutions were being stirred and titrated. This effect suggests that the final solutions were supersaturated with respect to NO_x . Despite the loss of NO_x during the titrations, however, the pH meter readings were not abnormally erratic.

4 SUMMARY

In this effort, we designed and tested a full-scale prototype of the nitric-acid dissolver system, which is capable of dissolving 250 g of irradiated LEU at ambient pressure for use in Mo-99 extraction. Three dissolution tests were performed:

- Dissolution of 197 g of depleted U to examine the extent of dissolution with time
- Dissolution of 202 g of depleted U to examine the self-heating of the dissolver solution by exothermic heat of dissolution.
- Dissolution of 242 g of depleted uranium plus 6.84 g of irradiated LEU to test the dissolver system at full scale and provide a feed solution for the Mo-99 extraction step.

All three tests were successfully completed. Tests results showed that the cooling system of the dissolver (reflux condenser) is sufficient to remove all heat produced by the exothermic dissolution reaction and the continuous decay heat of the irradiated foil. Results from these tests along with observations made during our manipulator mock-up test also demonstrated that our final design of the dissolver system is viable as a front-end process for the extraction of Mo-99 from high-density LEU foil targets.

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Chemical Sciences and Engineering Division

Argonne National Laboratory

9700 South Cass Avenue, Bldg. 205

Argonne, IL 60439-4837

www.anl.gov



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